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Report on the FINDER Experiment at the HIgS Facility

C. A. Hagmann, M. S. Johnson, D. P. McNabb, A. P. Tonchev, C.T. Angell, S. Hammond, C.R. Howell, A. Hutcheson, H.J. Karwowski, J.H. Kelley, E. Kwan, G. Rusev, W. Tornow

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C.A. Hagmann, M.S. Johnson, and D.P. McNabb
LLNL, Livermore CA, 94550
A.P. Tonchev, C.T. Angell, S. Hammond, C.R. Howell, A. Hutcheson, H.J. Karwowski, J.H. Kelley, E. Kwan, G. Rusev, and W. Tornow
TUNL, Durham NC, 27708

Introduction

Homeland Security programs are developing systems that use nuclear resonance fluorescence (NRF) to isotopically map a container. One such system being developed at LLNL is FINDER (Fluorescence Imaging in the Nuclear Domain with Extreme Radiation).

The proposed FINDER system works by impinging a tunable mono-energetic gamma ray beam onto a container under investigation. The photons pass through the container and a fraction of them scatter off of the interior components through various electromagnetic processes. One of these processes is NRF. At specific resonance energies, incident photons interact directly with the nuclei of special nuclear material (SNM) or other materials in the container. The incident beam is absorbed and scattered into all directions, depleting the spectrum at the resonant energy. The transmitted gamma ray beam accrues a notch a few eV in width after passing through the material of interest. This notched spectrum will impinge on a witness foil placed on the opposite side of the container relative to the gamma ray source. The witness foil will be made of material identical to the one being sought after. If there is a notch in the spectrum then there will be no NRF photons scattered from the witness foil. The corollary is that if there is no notch in the transmitted spectrum then there will be NRF photons scattered from the witness foil. A simple arrangement of gamma-ray detectors focused on the witness foil, are used to measure the NRF photons. If the detectors see NRF scatter then there was no NRF scatter within the container; therefore, no material of the nature being sought after was in that container. Conversely, if there was no NRF scatter from the witness foil, then the NRF scatter took place from within the container; therefore, the material of interest is inside of the container.

Recently, initial feasibility tests of FINDER was performed at the HIgS (High-Intensity Gamma Source) located at Duke University [1]. The preliminary results of these tests are discussed in this report. Our goals for these measurements were to demonstrate the concept of transmission detection and perform some initial validation of models of the FINDER concept. In particular, our models [2] indicated that backgrounds and nuisances are too small to obscure the high signal to noise of the FINDER technique. Therefore, FINDER offers extremely clear positive and negative signals for detecting SNM when measuring the high contrast attenuation of on-resonance gamma rays in transmission. Our initial demonstration of transmission detection provides a first check on our models – e.g. is there a physical process that we have forgotten to include? Also, previous work indicated that the notch could be obscured by small-angle scattering refilling the notch if the interrogating photon source is too broad in energy. While we did not expect notch refilling to be significant at HIgS which provides beam of gamma rays with ΔE/E ~ 3%, we were able to set an experimental upper limit on the amount of notch refilling in our experimental set up.

This report first describes the experimental setup for these measurements, gives a summary of the important results, and then concludes by discussing the extent to which we were able to validate our models [2] of the FINDER technique.

Experimental Setup

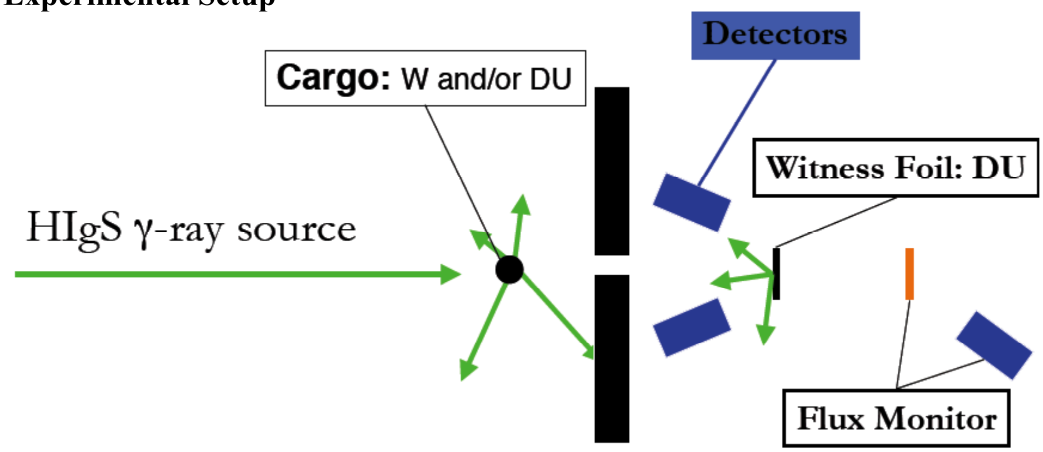

Figure 1: Schematic of FINDER feasibility test setup.

A schematic of the setup is shown in Figure 1. For these measurements cargo types of tungsten and depleted uranium were assembled. The thicknesses of the DU and W "cargos" were 1.27-cm thick. The cargo of DU and/or W was placed in front of a collimated wall with an opening of one inch. The different configurations are given in table 1. Depleted uranium was chosen to be the material of interest for this test, such that DU was placed in the witness foil location. The DU foil thickness was 3 mm and had a diameter of 2.54 cm.

Four HPGe detectors (60% efficiency of standard) were positioned at backward angles relative to the beam and facing the witness foil location (Fig 2). Two detectors were positioned at 100 degrees and the remaining two were positioned at 150 degrees. The detector to foil distance was 20 cm. Absorbers of 4 mm thick Cu were placed on the front face of the four detectors to reduce background. Sleeves of 2.54 cm thick Pb were placed around the HPGe's to reduce background from room scatter and other room sources.

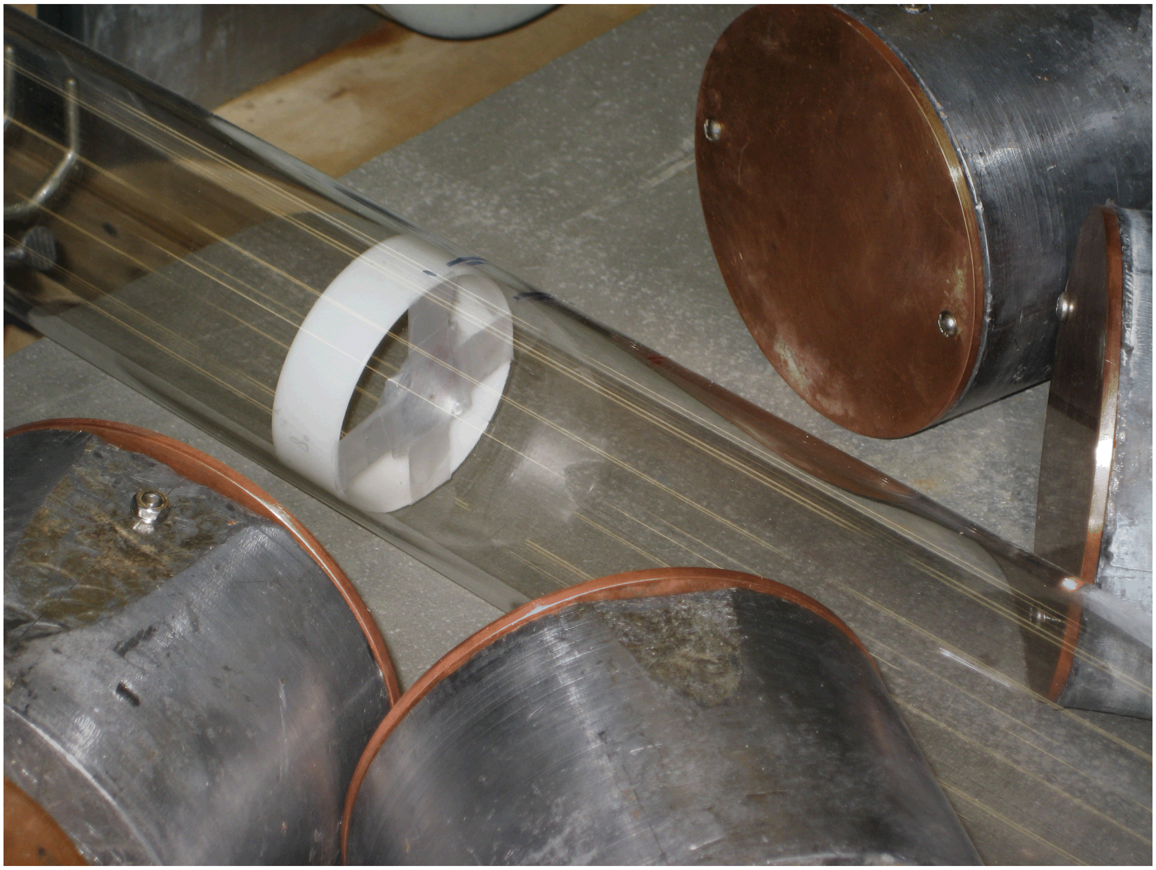

Figure 2: Target foil location. Beam direction from lower right to upper left. Four HPGes shown.

The flux from HIgS was monitored with a HPGe located downstream of the setup (Figures 1 and 4). Before each run the flux monitor was positioned on axis with the beam and the flux was measured directly. Figure 3 shows the measured incident spectrum, centered near 2180 keV and about 100 keV wide. Four 8” thick Cu attenuators were placed in the beam path far upstream during these short runs to reduce the dose to the flux monitor. Count rates corrected for the attenuation through 24” of copper result in an estimated peak flux 200 gammas/s/eV. During the production runs the flux monitor (HPGe) was rotated to 30 degrees relative to the beam and measured the Compton scattered flux from a 3 mm thick Cu foil as seen in Figure 4.

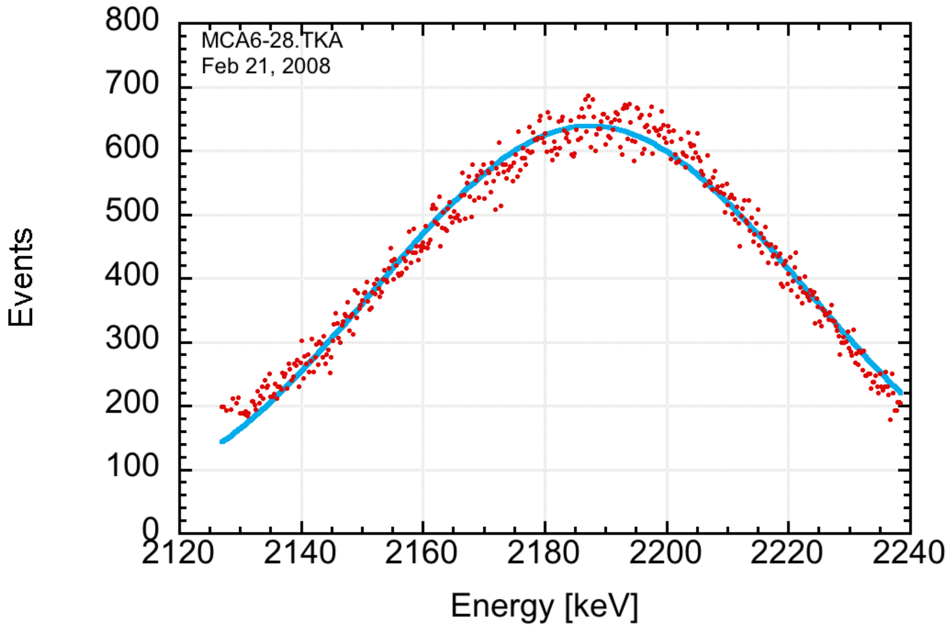

Figure 3 : Incident gamma spectrum measured with flux monitor located at 0 deg with respect to the beam.

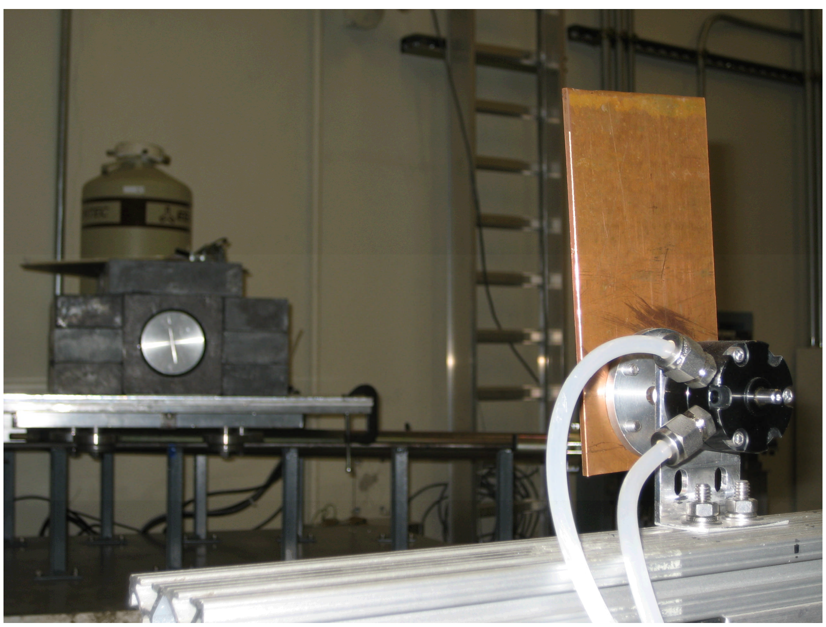

Figure 4: Flux Monitor. Gamma ray beam goes from right to left and impinges on the Cu plate (foreground). The Compton scatter is measured off-axis with an HPGe (background).

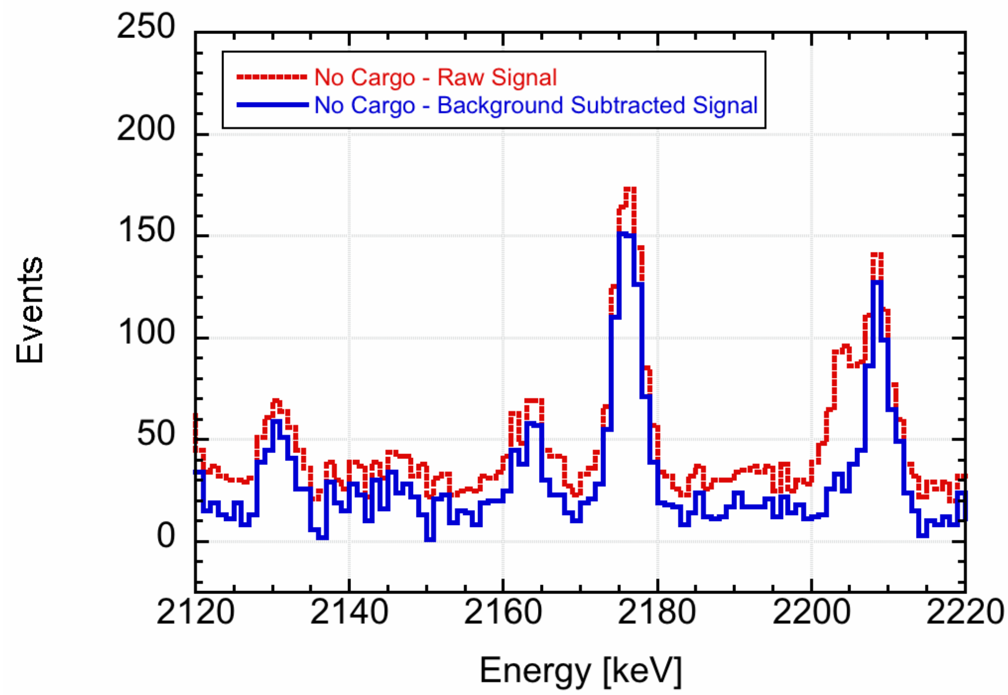

Figure 5: NRF spectrum of DU witness foil. The 2 peaks at 2176 and 2209 keV are due to direct transitions to the ground state in 238U. The smaller peaks at 2131 and 2163 keV are from decays to the first excited state in 238U at 45 keV. The baclground line at 2204 keV originates from beta decay of 214Bi.

Results

The goal of this experiment has been to demonstrate and validate the NRF notch detection concept and to explore potential pitfalls. For example, one known mechanism for filling the notch is Compton down-scattering of photons of energy above the NRF resonance producing the notch. Measurements on five different cargo configurations were made to explore these issues and the results are summarized in Table 1. The number of counts in the line at 2176 keV are given in raw form and normalized to the time-integrated flux that was recorded by the 30-deg flux monitor.

The first cargo scenario was "no cargo," which provides a useful data point for normalization and the energies and strengths of known 238U resonances to be verified. Figure 5 shows the observed spectrum of the "no cargo" measurement. The peaks at 2176 and 2209 keV are from M1 transitions between 1+ excited states and the 0+ ground state in 238U. The energies and ground state widths ($\Gamma_0 \sim 37$ meV) of both states have been previously reported by Heil et al. [3]. The smaller peaks at 2131 and 2163 keV are due to transitions to the first excited state in 238U. The line at 2204 keV arises from the decay of 214Bi which is part of the radioactive decay chain of 238U.

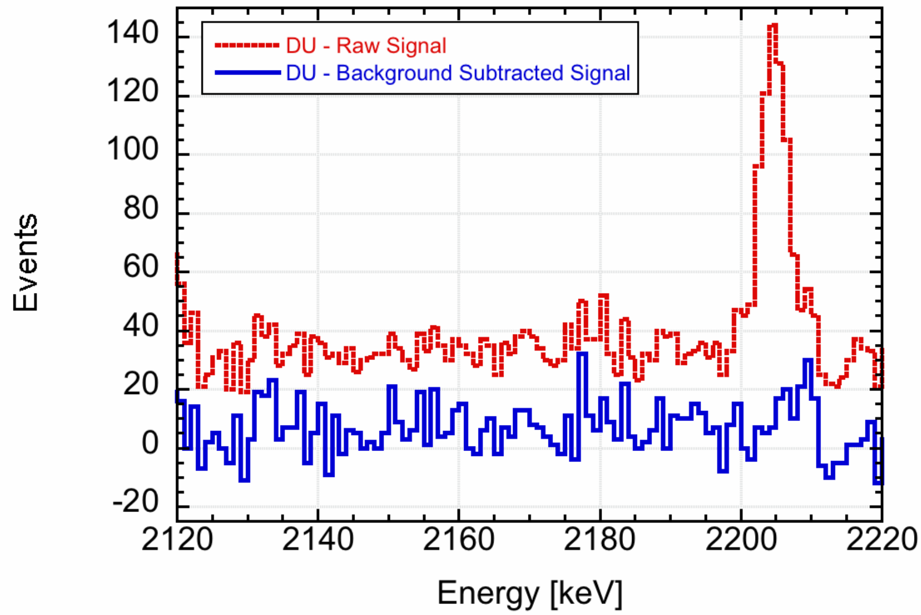

Figure 6: Measured spectrum with a ½" DU slab as the cargo.

Our second cargo scenario was a ½"-thick rectangular plate of depleted uranium. Figure 6 show the observed spectrum when the cargo is made of depleted uranium. As expected, the placement of 238U material upstream from the witness foil preferentially depleted the beam of photons at 2176- and 2209-keV resonance energies. Therefore, the amount of NRF scattering off the witness foil was substantially decreased relative to the total number of photons incident on the witness foil as measured by the flux monitor. The expected number of photons at the NRF resonance energy relative to nearby energies which only suffer attenuation due to atomic processes is simply $\exp(-x/x_{nrf})$, where x_{nrf} = 0.42 cm (37 meV/Γ_0)(19 g cm^{-3}/ ρ) is the NRF absorption length. The absorption length of both resonances in 238U of density 19.0 g/cm^3 is ~ 0.42(2) cm using the results from Heil et al. [3]. For our particular case of a ½"-thick slab of 238U, we expect a nuclear (NRF) attenuation of 0.050(3) on resonance relative to the intensity of non-resonant photons. The detailed shape of the notch is shown in Figure 7a for different values of Γ_0, assuming a NRF cross section thermally broadened to room temperature. Figure 7b shows the expected reduction in the number of NRF counts for a beam passing through ½" of U238 for a range of Γ_0 values.

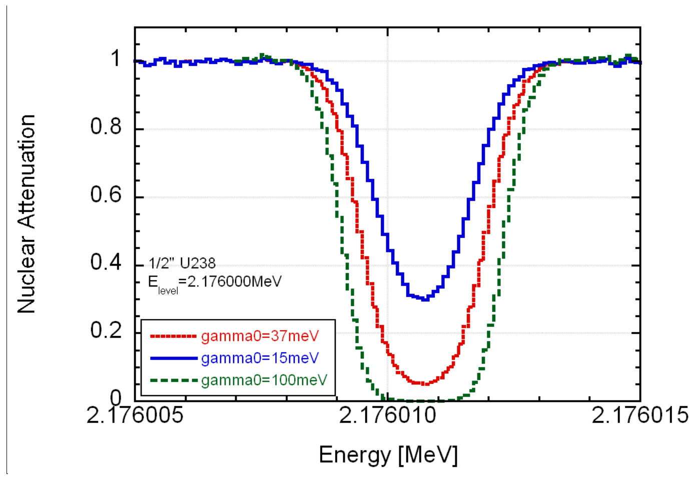

Figure 7a NRF absorption spectrum for a flat-top beam passing through a ½" slab of U238 (ρ =19 g cm^{-3}) at room temperature.

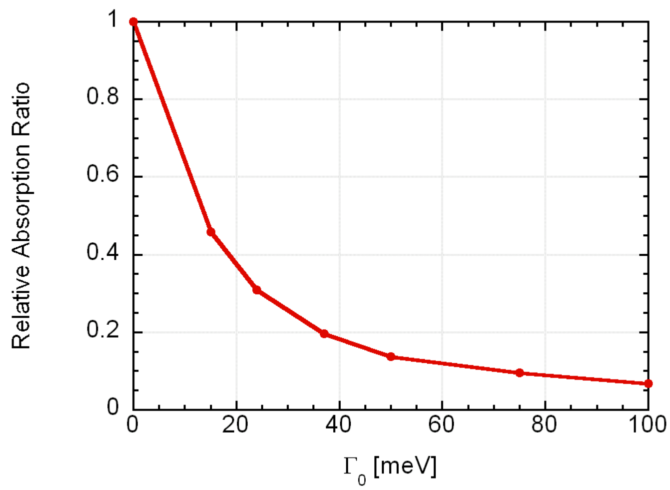

Figure 7b Ratio of NRF signal for the case of a ½" slab of U238 in front of the witness foil to the 'No Cargo' case.

Our third cargo scenario was a ½"-thick rectangular plate of tungsten. We chose tungsten because the density of tungsten and uranium are very similar (19.3 versus 19.0) and are essentially indistinguishable from each other in a conventional radiograph. The spectrum obtained is shown in Figure 8. The photons on resonance are not depleted, and results are consistent with no nuclear attenuation.

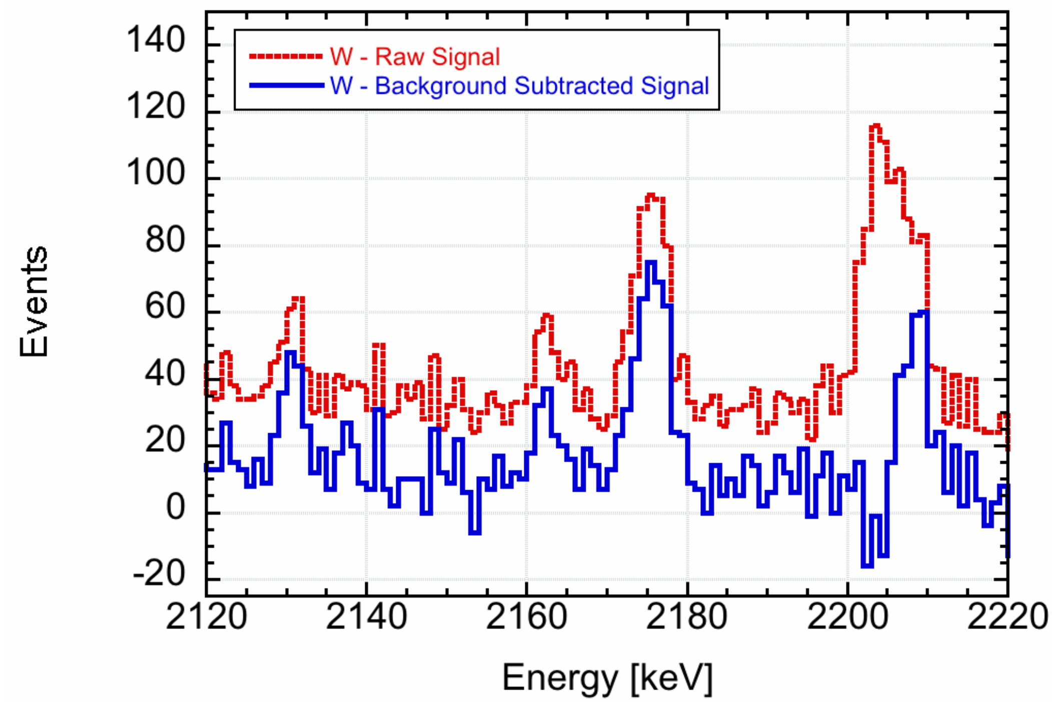

Figure 8: Measured spectrum with a ½" W slab as the cargo.

In order to explore the possibility of small angle scattering obfuscating the notch in the transmitted spectrum, we took data for two other "mixed" cargo scenarios with both tungsten and uranium as cargo. First, the ½" tungsten plate was placed upstream of the ½" depleted uranium plate. In this case the tungsten cannot cause the notch to be refilled and so the nuclear attenuation should be consistent with the uranium-only scenario. In the other case, we placed the tungsten plate downstream of the uranium plate. The results are clearly consistent with each other – in other words the data are consistent with no notch refilling.

Table 1: Different cargo configurations listed on left. Raw counts and counts normalized to fluence in the peak at 2176 keV are shown.

Cargo Type	Meas. Time [hrs]	Normalized flux at Witness foil	Raw counts	Flux-normalized counts	Relative Absorption Ratio
No cargo	2.7	1	633 (30)	633 (30)	1
½" DU	9.3	0.694	7 (21)	10 (30)	0.02 (5)
½" W	4.0	0.538	349 (27)	649 (50)	1.03 (9)
½" DU + ½" W	25.5	0.836	141 (44)	169 (53)	0.27 (8)
½" W + ½" DU	24.3	0.901	129 (46)	143 (51)	0.23 (8)

Table 2: Different cargo configurations listed on left. Raw counts and counts normalized to fluence in the peak at 2209 keV are shown.

Cargo Type	Meas. Time [hrs]	Normalized flux at Witness foil	Raw counts	Flux-normalized counts	Relative Absorption Ratio
No cargo	2.7	1	444 (24)	444 (24)	1
½" DU	9.3	0.694	75 (23)	108 (33)	0.24 (8)
½" W	4.0	0.538	224 (16)	416 (30)	0.94 (8)
½" DU + ½" W	25.5	0.836	110 (52)	132 (62)	0.30 (14)
½" W + ½" DU	24.3	0.901	125 (52)	139 (58)	0.31 (13)

Discussion

The measurements discussed in this report clearly demonstrate that transmission-based, isotopic detection works and can distinguish different elements from each other, even if they have the same areal density. Specifically, the FINDER technique was able to distinguish the uranium plate from the tungsten plate even though they have the same areal density. Equation 27 of Reference [2] reveals that six-sigma detection of ½" of uranium requires 50 counts. Based on the results of Table 1, this corresponds to about 6(2) hours of beam time at HIGS. In order to reduce the count time from 6 hours to 1 second, the flux would have to be increased from ~200 /eV/s to ~4×10^6 /eV/s.

Beyond demonstrating the FINDER technique, the measurements presented here do help to validate the background and count rate models published in [2]. In [2], the authors focus on very short count times and the dominate backgrounds in the notch detectors placed around the witness foil arise from elastic and multi-step scattering of the beam. In these hour-long measurements, however, the dominate background arises from radioactive decay of 238U atoms in the witness foil and other room background. Clearly, this background term will not be present in high-flux measurements. On the other hand, beam-correlated backgrounds (elastic scattering and bremsstrahlung) are expected to persist. At energies within a few percent of the beam energy, elastic processes (Rayleigh, nuclear Thomson, Delbrück, and GDR scattering) should be dominant. To estimate the elastic scatter term, we fitted the background subtracted 'No-cargo' spectrum with the Gaussian spectrum function depicted in Figure 3. The results are shown in Figure 9. Using the known cross section and angular distribution [4] of the NRF radiation for calibration and correcting for the finite thickness of the U238 witness foil, we estimate an elastic scattering cross section (polarization parallel to the scattering plane) $d\sigma_{el}/d\Omega$ = 79±13 μb/sr at θ ~120°, which is in agreement with the previous theoretical estimate of $d\sigma_{el}/d\Omega$ ~ 70 μb/sr [5].

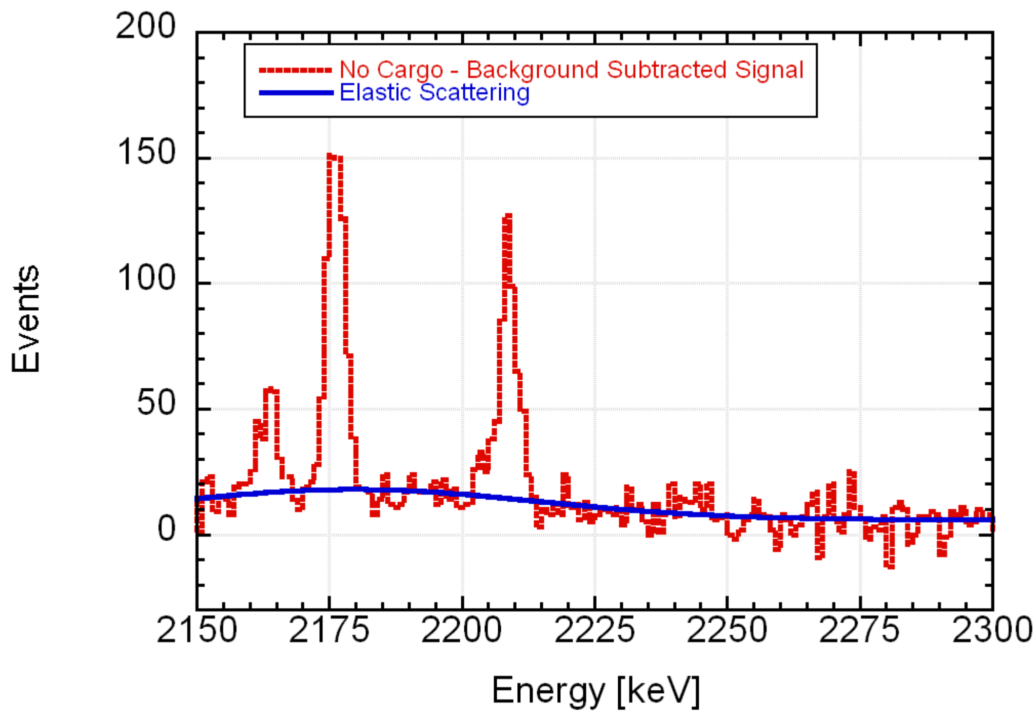

Figure 9 The red curve shows the 'No Cargo' spectrum with beam-uncorrelated background subtracted. The blue curve represents the continuous beam-correlated background.

It is also important to validate that the notch used for detection in the FINDER technique is not filled in by small-angle scattering processes through thick amounts of shielding.

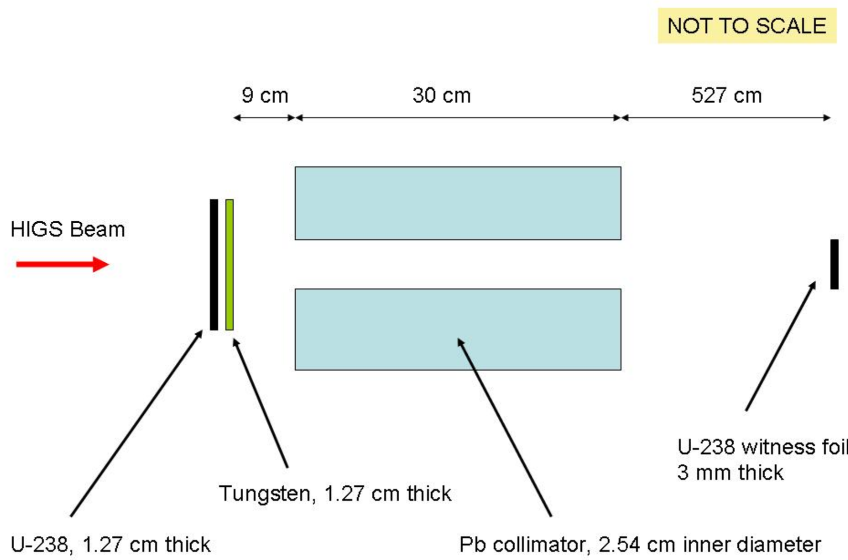

Figure 10: Geometric configuration of NRF notch fill measurement

The expected amount of notch filling (~ 10^{-7}) was computed with MCNP for the geometry shown in Figure 10. In the simulation, a perfect notch (3 keV wide) was placed at 2176 keV and the measured gamma spectrum shown in Figure 3 was used for the source particles. In the present experiment, the U-238 witness foil subtends a very small solid angle at the location of the W foil. In the paper by Pruet et al. [2], the witness foil was placed adjacent to the scattering material and larger filling fractions were predicted.

For these measurements, the amount of notch refilling can also be written as

$$f = \frac{N_{U+W} - N_U}{N_{\text{no cargo}} - N_U}.$$

In this expression, N stands for the flux normalized counts which is proportional to the depth of the notch. N_{U+W}-N_U represents the number of photons scattered into the depleted notch, and $N_{\text{no cargo}}$-N_U represents the depth of the notch as it exits the U plate. Combining the counts from Tables 1 and 2, result in f = 0.22(9), which is consistent with zero. It is hard to make a substantially better measurement of the notch refilling fraction without a substantially larger (~100X) flux or a different measurement geometry.

If we assume that notch-refilling is negligible, the three measurements of the nuclear attenuation factor with the ½" uranium plate also represent an independent, direct measure of the resonance width of the states in 238U. For the 2176-keV resonance, the weighted-average relative absorption ratio is 0.12(4), which corresponds to a resonance width of 67(16) meV. For the 2209-keV resonance, the weighted-average relative absorption ratio is 0.27(6), which corresponds to a resonance width of 28(8) meV. In both cases, these widths are in rough agreement with the 37(2) meV reported by Heil et al. [3].

Conclusions

We demonstrated that transmission-based, isotopic detection works and can distinguish different elements from each other, even if they have the same areal density. Specifically, the FINDER technique was able to distinguish the uranium plate from the tungsten plate even though they have the same areal density. Our models for backgrounds and notch refilling were also validated, but it was also shown that ~100 times more flux is needed before a detailed comparison with model calculations of the dominate notch-refilling terms can be made. Future work at these fluxes should focus on validating detection algorithms and comparing reflections and transmission-based detection.

References

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